Magnetic Relaxation of Permeability (Disaccommodation) in an Amorphous Ferromagnetic Fe-Si-B-C Alloy

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Abstract: Disaccommodation (*DA*) was examined in the temperature range from -196°C to the Curie temperature for an amorphous $Fe_{78.4}$ $Si_{8.6}$ $B_{12.7}$ $C_{0.3}$ alloy, which exhibits a maximum around 250°C. When the alloy is annealed successively at various temperatures below 220°C, the *DA* vs *T* curve does not change. On the other hand, it shifts to higher temperatures when the alloy is annealed above 220°C. The temperature dependence of the *DA* and magnetic anisotropy is explained by the model of compositional short-range ordering proposed by Beukel and Radelaar.

1. Introduction

Amorphous Fe-Si-B-based alloys exhibit a relatively high saturation induction and low core loss [1] and are expected to have uses as materials for commercial frequency or medium frequency (1-100 kHz) transformers. Practical use of amorphous material requires magnetic and mechanical stability, in addition to their soft magnetic properties. On the other hand, an-Fe-based amorphous alloy exhibits features that are different from Co-based amorphous alloys with regard to magnetic relaxation [2, 3]. Therefore, it is of interest to study the magnetic relaxation for Fe-based amorphous alloys. In spite of numerous works concerning the magnetic relaxation of Co-based and Fe-Ni-based amorphous alloys [4-10], little has been studied about the Fe-Si-B-based amorphous alloys.

The present paper describes the temperature dependence of the time decrease of initial permeability (disaccommodation, *DA*) in the temperature range -196°C to the Curie temperature for an amorphous $Fe_{78.4}$ $Si_{8.6}$ $B_{12.7}$ $C_{0.3}$ alloy and discusses the origin of *DA*.

2. Experiment

An amorphous $Fe_{78.4}$ $Si_{8.6}$ $B_{12.7}$ $C_{0.3}$ alloy was produced in a ribbon form (25mm wide, 20-25 μm thick) using a rapid quenching disk method. The structural analysis of the ribbon sample was carried out by the Debye-Scherrer X-ray technique. The diffraction patterns exhibited a diffused halo.

The samples for the measurement of permeability μ were prepared by the same method as was reported previously [9]. First, the ribbon was cut into a section one meter long and 5 mm wide and wound around a ceramic ring with a diameter of 15 mm. The insulation between each of the layers was formed from an Al_2O_3 powder. An insulated copper wire of 0.3 mm in diameter was wound around the core ring in 220 turn primary coil and 110 turn secondary coil.

Permeability was measured by alternating a driving magnetic field (140-9200 Hz, 0-0.25 Oe, time period : 4 min) with demagnetization (50 Hz, 50 Oe, time period : 2 sec)

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using an automatic *DA* measuring apparatus. For the measurement of the temperature dependence of μ_i, the sample was heated and/or cooled at a rate of about 60°C/h.

The magnetic anisotropy was measured using a torque magnetometer and a disk-shaped sample. The sample was heated and/or cooled at a rate of about 100°C/h.

3. Experimental result

3. 1 Dependence of μ and DA on the driving field at room temperature

Figure. 1 shows the dependence of μ and *DA* on the driving field under the conditions of several different fixed frequencies at room temperature. Here the magnitude of *DA* is defined as $DA\ (\%) = [\mu_i(0) - \mu_i(4)\}/\mu_i(0)] \times 100$, where $\mu_i(0)$ and $\mu_i(4)$ are the initial permeabilities at 0 and 4 min after demagnetization, respectively. The μ is nearly constant up to 50 mOe and increases gradually with an increasing driving field. *DA* is observed in the initial permeability range exhibiting a maximum at about 15 mOe. The curve of *DA* versus driving field is similar to the bell shaped curve observed in crystalline alloys [11] which is related to the after-effect caused by interstitial diffusion or reorientation of atomic pairs of constituted atoms.

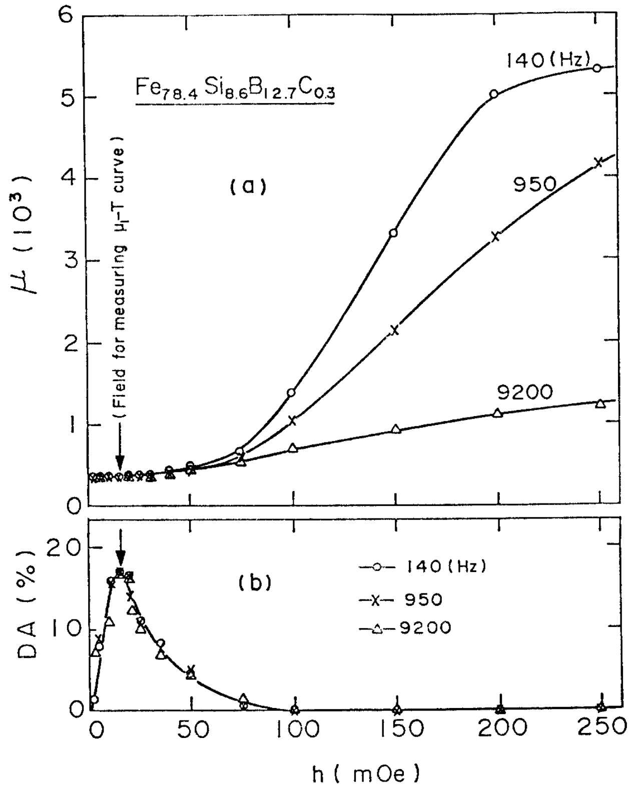

Fig. 1 Dependence of μ (a) and *DA* (b) on driving field.

The data mentioned hereafter were measured at the driving field indicated by the arrows in the figure, since the magnitude corresponde to the initial permeability range and the *DA* is so large that we can measure it easily.

3. 2 Temperature dependende of μ_i and DA

Figure. 2 shows the temperature dependence of μ_i and *DA* measured by alternating demagnetiztion and application of a driving field. As seen in the figure, μ_i exhibits a broad maximum around room temperature during heating, and a sharp large maximum during cooling, which contrasts with the case of the amorphous $Fe_5Co_{70}Si_{15}B_{10}$ alloy[12]. The *DA* exhibits a large maximum at about 250°C on heating and a broad one at about 150°C on cooling.

In order to make clear the influence of the structural relaxation upon the *DA*, the sample was annealed successively at higher temperatures those in the measurement of *DA* for about 15 min. As can be clearly seen in fig. 3, the annealing temperature being lower than 220°C, the *DA* vs. *T* curve is reversible. But it becomes irreversible and shifts to higher temperatures as the annealing temperature is increased higher than 250°C. This trend was also observed in a binary amorphous $Fe_{85}B_{15}$ alloy [15]. When the annealing temperature is raised above 400°C, the *DA* vs. *T* curve shifts to a lower temperature at about 200°C and the maximum becomes flattened.

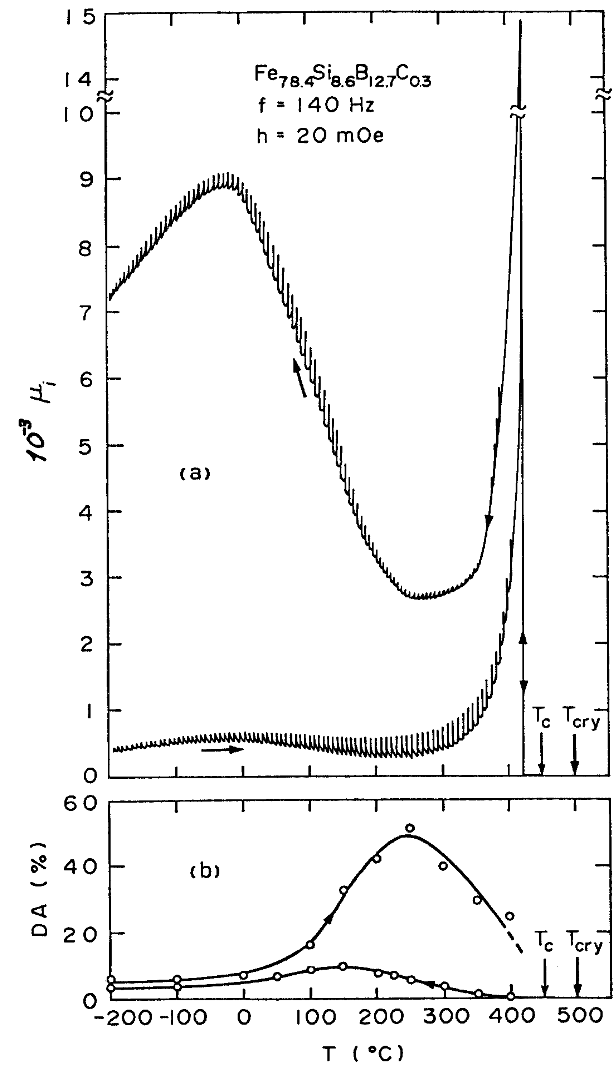

Fig. 2 Temperature dependence of μ_i (a) and DA (b).
(Tc: Curie temperature, $Tcry$ = Crystalization temperature)

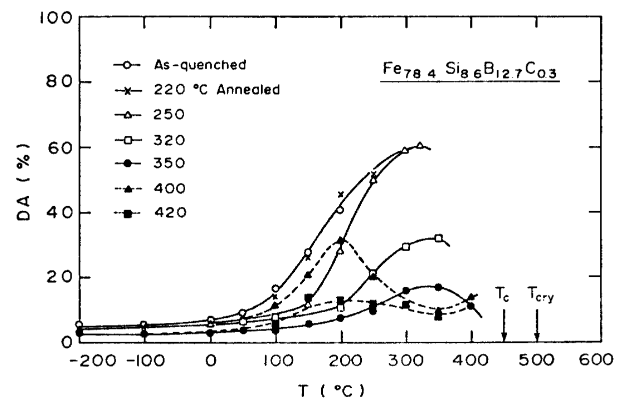

Fig. 3 Temperature dependence of DA after annealing at various temperatures.

3. 3 Time dependence of μ_i

In order to obtain a further insight into the relation between structural relaxation and DA, the measurement of DA was carried out at various temperatures below 200°C for a long time up to several tens of hours. Figure 4 shows the time dependence of μ_i along with the $\mu_i(t)/\mu_i(t\to\infty)$ vs. ln t plot. As can be seen in the figure, μ_i continues to decrease gradually over the several tens of hours below 142°C. When the temperature is increased further, μ_i decreases faster and saturates in a several tens of min. at 200°C. Here it should be noted that $\mu_i(t)/\mu_i(t\to\infty)$ is proportional to ln t during the intermediate measuring time. This may be due to the distribution of the activation energy.

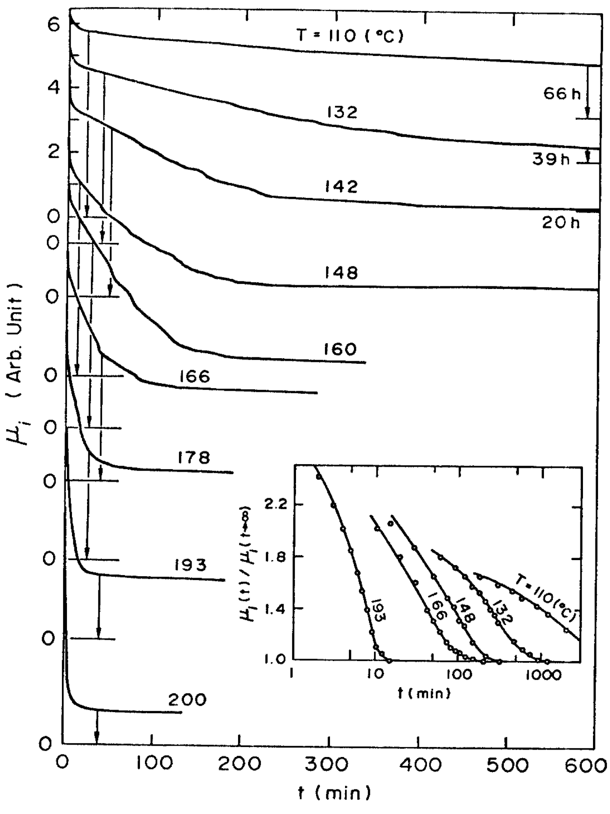

Fig. 4 Time dependence of μ_i. Inset shows $\mu_i(t)/\mu_i(t\to\infty)$ vs. ln t plot.

In order to obtain an activation energy, the relaxation time τ is obtained from the data in fig. 4 and plotted as a function of $1/T$ in fig. 5. In this figure, the results for $Fe_{85}B_{15}$ and $Fe_{75}B_{25}$ alloys are also shown. The activation energies E_a evaluated from the slope of the straight lines are 0.97, 0,21 and 0.37 eV for $Fe_{78.4}Si_{8.6}B_{12.7}C_{0.3}$, $Fe_{85}B_{15}$

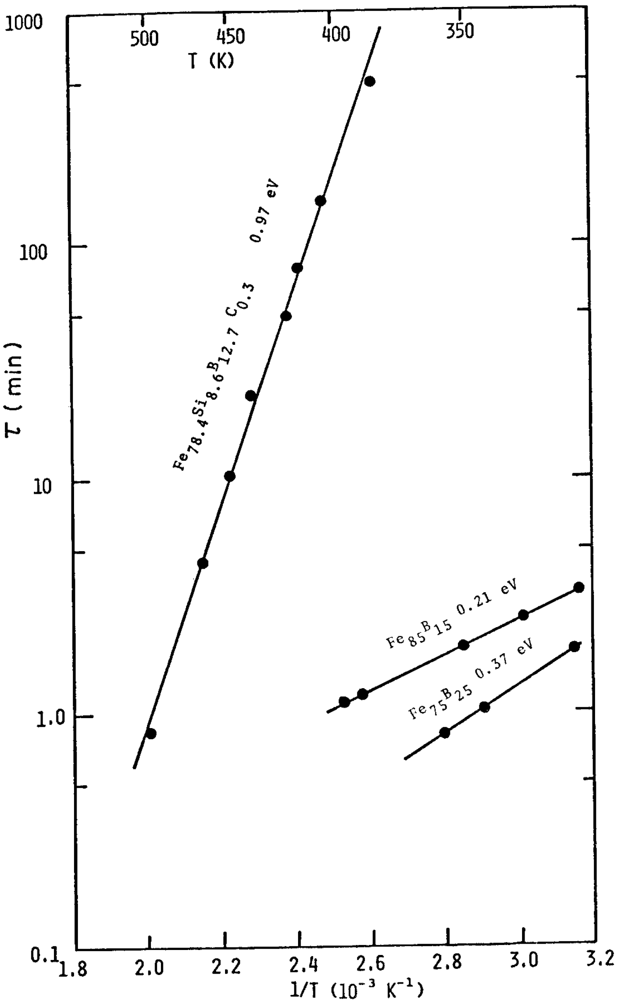

Fig. 5 ln τ vs. $1/T$ plot.

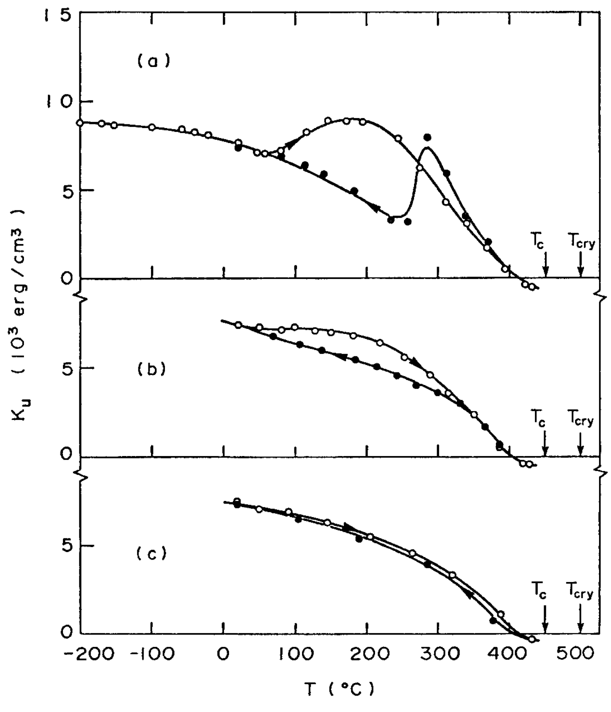

Fig. 6 Temperature dependence of uniaxial magnetic anisotropy constant. 1st cycle (a), 2nd cycle (b) and 3rd cycle (c).

and $Fe_{75}B_{25}$, respectively. It should be noted that E_a does not depend on the B content. On the other hand the E_a of $Fe_{78.4}Si_{8.6}B_{12.7}C_{0.3}$ alloy is more than 2.7 times larger than that of Fe-B alloys.

3. 4 Temperature dependence of magnetic anisotropy

Figure 6 shows the temperature dependence of the magnetic anisotropy constant K_u measured through repeated heating and cooling. The K_u changes in an irreversible manner in the temperature range from 75 to 300°C. It should be noted that K_u exhibits a broad maximum at about 200°C during heating and it becomes broader through repetition of the heating cycle. Furthermore, the temperature roughly corresponds to the temperature of the maximum in the DA-T curve (see fig. 2 (b)).

4. Discussion

The origin of DA in amorphous alloys has been discussed in terms of the two mechanisms depending upon temperature. Allia et al. [13] have explained it by the magnetostrictive coupling between structural defects and saturation magnetization for iron-based amorphous alloys at room temperature. On the other hand, Kronmüller [7, 8] and the authors of this paper [9] have explained the DA above room temperature by directional ordering of constituted atoms for Fe-Ni-based amorphous alloys. However, no detailed explanation of the DA vs T curves has been reported taking into account the structural relaxation of amorphous alloys.

Recently, Beukel and Radelaar [14] have calculated the change of free volume v_f and the order parameter α during isochronal annealing for a binary amorphous alloy. Even if the calculation is based on the phenomenological concept of compositional short-range ordering, the results explain some changes in physical properties caused by the structural relaxation. The difference in the order parameter $\Delta\alpha$ between at room temperature (as quenched state) and at higher temperature is shown in fig. 7 together with the result concerning free volume [14]. As can be seen in fig. 7 (b), $\alpha\Delta$ exhibits a maximum and the $\Delta\alpha$ vs. T curve shifts to higher temperatures in accordance with decreases in the cooling rate, i. e. the decreases in free volume. Here, it should be noted that the temperature dependence of *DA* measured after annealing at various temperatures (see fig. 3) is similar to the $\Delta\alpha$ vs. T curves. Furthermore, $\Delta\alpha$ increases in the low temperature region where v_f is nearly constant.

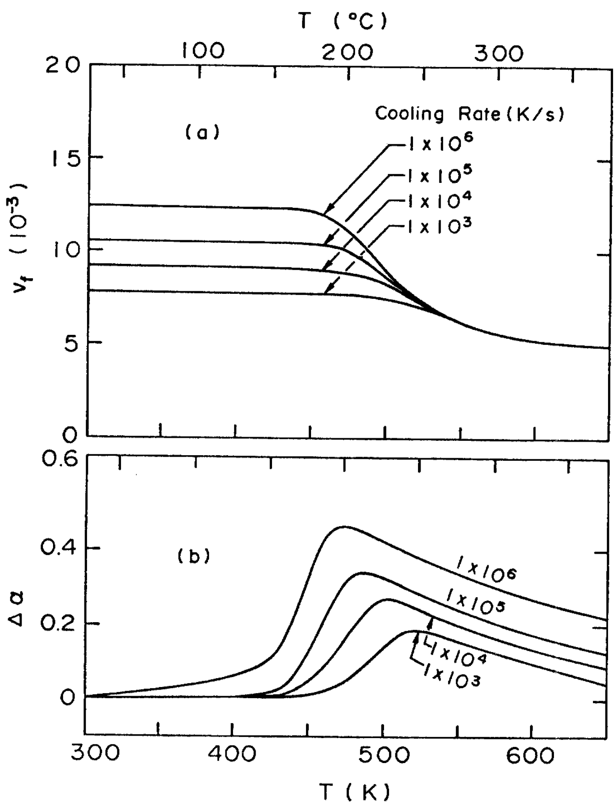

Fig. 7 Change of the free volum v_f (a) and the order parameter $\Delta\alpha$ (b) during isochronal annealing (heating rate 0.33 K/s) of as-quenched A-B binary alloys. The result was calculated by Beukel and Radelaar [14].

According to the recent experiment concerning the length changes done by Jagielinski and Egami [15], the volume change is always irreversible and monotonic. From these facts taken together, it would be expected that the physical properties reflecting the compositional short range ordering changes are reversible at low temperatures where v_f is constant, with changes that are irreversible at higher temperatures where v_f decreases. The *DA* measured in the present study can be interpreted as a reflection of the compositional short-range ordering. Recently, we have examined the compositional dependence of DA for amorphous Fe-B and Co-B alloys in the temperature range from -196°C to about 300°C[16]. The compositional dependence of *DA* below room temperatures was almost the same as that above room temperature. The results suggest that the origin of *DA* below room temperature should be ascribed to be the same. Therefore, we interpret the temperature dependence of *DA* as follows.

When the temperature is low enough to prevent a decrease in excess free volume, subtle change of constituent atoms may take place by means of the migration of free volume and the atom pairs may reorient their axis, corresponding to an increase in the order parameter. These atom pairs induce a local magnetic anisotropy to decrease the μ_i. When the temperature is raised above room temperature, the excess free volume is annihilated to some degree and atom pairs are frozen causing an irreversible decrease in μ_i.

5. Summary

The *DA* and magnetic anisotropy were measured in the temperature range from -196°C to the Curie temperature for an amorphous $Fe_{78.4}Si_{8.6}B_{12.7}Co_{0.3}$ alloy. The results obtained are summarized as follows.

(i) The DA exhibits the maximum (about 50%) around 250°C.

(ii) The magnetic anisotropy also exhibits a broad maximum around 200°C, which corresponds to an increase in compositional short-range ordering.

(iii) When the alloy is annealed successively at various temperatures below 220°C, the DA vs. T curve does not change. On the other hand, it shifts to higher temperatures when the alloy is annealed above 220°C.

(iv) The activation energy of the DA is 0.97 eV.

(v) The DA is explained by the model of compositional short-range ordering proposed by Beukel and Radelaar.

References

[1] R. Hasegawa: J. Magn. Magn. Mater. 41, 79 (1984).

[2] T. Miyazaki, M. Takahashi, Y. H. Lee, K. Hisatake and T. Yamamoto: Proc. Int. Conf. Rapidly Quenched Metals, Sendai p. 1179 1981 (Japan. Inst. Metals).

[3] T. Miyazaki and M. Takahashi: unpublished.

[4] T. Matsuyama, K. Ohta, M. Kajiura and T. Teranishi: Japan. J. Appl. Phys. 19, 55 (1980)

[5] E. Kisdi-Koszo, P. Vojtanik and L. Potocky: J. Magn. Magn. Mater. 19, 159 (1980).

[6] T. Egami: Mat. Res. Bull. 13, 557 (1978).

[7] N. Moser and H. Kronmüller: J. Magn. Magn. Mater. 19, 275 (1980).

[8] H. Kronmüller: J. Magn. Magn. Mater. 41, 366 (1984).

[9] T. Miyazaki, K. Hisatake and M. Takahashi: Japan. J. Appl. Phys. 22, 1277 (1983).

[10] H. Börner and R. Höhne: phys. stat. sol. 76, 499 (1983).

[11] L. Neel: J. Phys. Radium 13, 249 (1952).

[12] T. Miyazaki and M. Takahashi: Sci. Rep. RITU, Suppl. A 28, 311 (1980).

[13] P. Allia and F. Vinai: Phys. Rev. B 26, 6141 (1982).

[14] A. V. D. Beukel and S. Radelaar: Acta Metal. 31, 419 (1983).

[15] T. Miyazaki, M. Takahashi and K. Hisatake: J. Appl. Phys. 57, 3575 (1985).